

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of

**van BENTHEM et al**

Atty. Ref.: 4662-131

Serial No: 10/565,963

Group: 1796

Filed: February 28, 2006

Examiner: Dollinger

For: **ADHESIVE COMPOSITION COMPRISING A FORMALDEHYDE-CONTAINING AMINOPLAST RESIN AND A CATALYSING COMPOUND**

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Commissioner for Patents  
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FACTUAL DECLARATION UNDER 37 CFR §1.132

Sir:

Pursuant to 37 CFR §1.132, the undersigned, Karel F.H. BONEKAMP, hereby declares and states:

1. I am a named co-inventor of the subject matter disclosed and claimed in the above-identified US patent application ("the '963 application").
2. On information and belief, I understand that the US Patent Examiner has cited EP 0 107 260 A1 (Tinkelenberg et al) to reject several of the pending claims in the '963 application. The purpose of this Factual Declaration is to rebut the Examiner's rationale for applying Tinkelenberg et al as a reference against such pending claims.
3. Tinkelenberg et al defines a catalyzed melamine-urea-formaldehyde (MUF) resin a F/NH<sub>2</sub> molar ratio of 0.25 – 0.625 which is equivalent to a ratio F/(NH<sub>2</sub>)<sub>2</sub> of 0.50 - 1.25). The catalyst of Tinkelenberg et al comprises a mixture of latent catalyst

and a strong acid in an effective amount. Furthermore it is defined in claim 2 of Tinkelenberg that a sufficient amount of strong acid is to be used for completing the pressing process at 180°C within 10 seconds per mm sheet thickness. This amount of strong acid is shown in example III to be 7.5 wt.% of 4N HCl solution apart from the 1.5 wt.% dry NH<sub>4</sub>Cl latent catalyst, both percentages calculated on the basis of dry adhesive. The catalyst amount in Tinkelenberg et al is needed for an adhesive system which does not contain MDI as second component.

4. In the '963 application, an adhesive having a pH of lower than 7.0 is defined, with a preferred more specific adhesive pH range of 6.5 – 5.5. As will become clear from the evidence presented below, the pH range applied in Tinkelenberg et al is completely different from the pH range employed in the invention claimed in the '963 application.
5. The following experimental procedures were performed under my direction and control:
  - a. **Adhesive resin preparation**

Tinkelenberg et al show in example III (table II), that upon using 7.5 wt.% of 4N HCl solution and 1.5 wt.% dry NH<sub>4</sub>Cl (both percentages calculated on dry adhesive resin), panels can be prepared with good bonding and emission properties.

The adhesive resin of example III is composed of following constituents:

- 1) MF resin with F/M molar ratio of 1.65
- 2) UF resin with F/U molar ratio of 1.25
- 3) Urea

The final adhesive resin is specified in Tinkelenberg et al as follows: F/NH<sub>2</sub> = 0.5 (so F/(NH<sub>2</sub>)<sub>2</sub> = 1.0) and the melamine content is 7.7 wt.% on dry adhesive resin. Solid Content (SC) is not specified.

The adhesive resin as disclosed by Tinkelenberg et al example III (table II) was repeated according the same final specifications as mentioned therein. Specifically, the repeated experimental example which was prepared included the following constituents:

- 1) MF resin with F/M molar ratio of 1.65 and calculated SC of 55%
- 2) UFC 80, which is a stable UF pre-condensate with F/U molar ratio of 5 and calculated solid content of 80% (readily available commercial product)
- 3) Urea

The components specified immediately above were used in following weight ratio: 1) MF resin, 100 parts; 2) UFC 80, 229 parts and 3) Urea, 211 parts.

Upon mixing the constituents and dissolving urea at 40°C, a clear adhesive resin was obtained after two hours with F/(NH<sub>2</sub>)<sub>2</sub> = 1.0 and a melamine content of 7.7 wt.% on dry adhesive resin. After 24 hours the formaldehyde smell had disappeared, indicating that free formaldehyde had been reacted into the system. The pH was adjusted to 9.0 with NaOH solution before titration. This is a normal setting used for the purpose of adhesive resin stability. Calculated SC was 83%, practical SC (determined after weight loss for 2 hours at 120°C) was 70%, which is common practice for such an adhesive resin.

The adhesive resin of the repeated example now had essentially the same properties as the adhesive resin as disclosed in Tinkelenberg et al. It is known from my experience in this field, that final properties of the adhesive resin depend on F/(NH<sub>2</sub>)<sub>2</sub> molar ratio and melamine content. This is certainly the case for pH

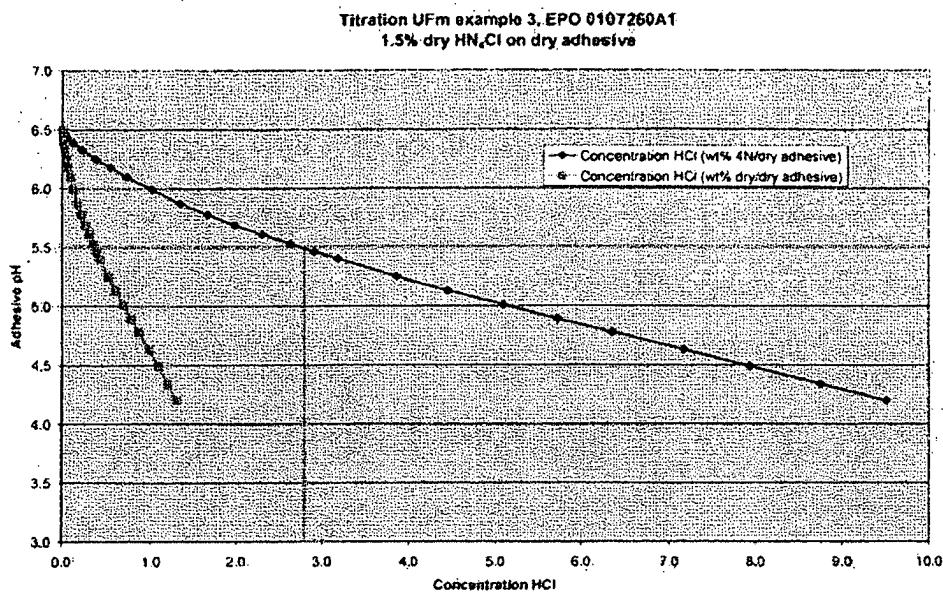
properties. The reason for using UFC80 instead of F/U 1.25 resin is that it is available as a stable commercial product. Moreover, it is fairly possible that the UF resin, used in Tinkelenberg et al was also originally prepared from UFC80 and urea, as this is often the case.

b. Titration

The above prepared adhesive resin was catalyzed with 1.5 wt.% dry NH<sub>4</sub>Cl (latent catalyst) on a dry adhesive basis and the pH was measured at 22°C with a ORION 8172BNWP sleeve electrode combined with a Mettler DL25 titrating / pH measuring device.

After adding the NH<sub>4</sub>Cl, the pH started decreasing slowly, because of a reaction of the ammonia with free formaldehyde, resulting in a release of HCl. Because the free formaldehyde had to be extracted from the adhesive, this process took some time. The liberation of the acid was therefore delayed, meaning a longer pot-life, which is the function of this latent catalyst. After ½ hour the pH was lowered from 9.0 till 6.8 and after 1 hour the pH was 6.5 and did not further decrease significantly.

Titration with 4 N HCl was started subsequently and the pH results were compiled in graph 1 below.



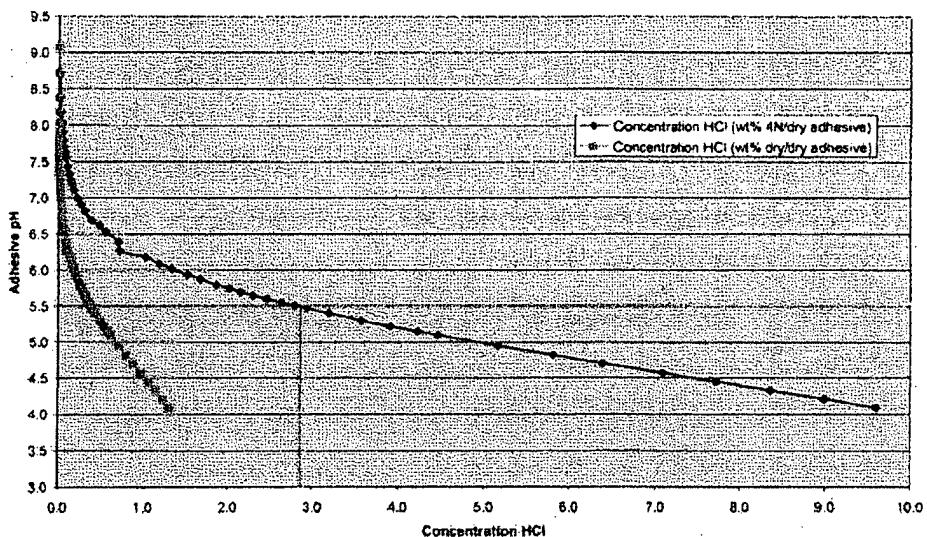
Graph 1. Titration of adhesive with additional  $\text{NH}_4\text{Cl}$  latent catalyst, start 1 hour after adding  $\text{NH}_4\text{Cl}$

2.7 Wt. % of 4N HCl solution on dry adhesive was needed to reach pH 5.5. This equals 0.4 wt. % dry HCl on dry adhesive. With 7.5 wt. % of 4N HCl solution on a dry adhesive basis like that employed in Example III of Tinkelenberg et al, the pH is lowered further until around 4.6.

To look if additional pH effects occurred with  $\text{NH}_4\text{Cl}$  in this low pH region, a titration without  $\text{NH}_4\text{Cl}$  was also performed. The reason for doing this was that it might be possible that some buffering effect resulted from the reaction products from ammonia and free formaldehyde. This was however expected not to have a major influence on the pH.

Nevertheless, in graph 2, titration results without  $\text{NH}_4\text{Cl}$  are presented.

Titration UFm example 3, EPO 0107260A1.  
No additional  $\text{NH}_4\text{Cl}$  in the adhesive



Graph 2. Titration of adhesive not containing additional  $\text{NH}_4\text{Cl}$  latent catalyst

With no  $\text{NH}_4\text{Cl}$  being added, pH 5.5 was reached with 2.8 wt. % of 4N HCl solution on a dry adhesive basis. This equals 0.4 wt. % dry HCl on dry adhesive.

When using only HCl in the amount as used in Tinkelenberg et al (7.5 wt. % of 4N HCl solution on dry adhesive), adhesive pH is around 4.5 which is in the same region as with  $\text{NH}_4\text{Cl}$ . As expected, the influence of  $\text{NH}_4\text{Cl}$  on adhesive pH was not significant when using these higher amounts of HCl.

van BENTHEM et al  
Serial No. 10/565,963

6. Based on the evidence provided above, the following conclusions can be made:

- In the adhesive composition according Tinkelenberg et al example III (with NH<sub>4</sub>Cl), an adhesive pH of 5.5 was reached with an amount of 2.7 wt. % of 4N HCl solution on dry adhesive (0.4 wt. % dry HCl on dry adhesive). At this concentration HCl, the presence of 1.5 wt. % NH<sub>4</sub>Cl dry on dry adhesive has minor additional influence on pH (see results in graph 1 and graph 2).
- 7.5 Wt. % of 4N HCl solution on dry adhesive, as used in Tinkelenberg et al example III (with NH<sub>4</sub>Cl) resulted in an adhesive pH of around 4.6, far below 5.5 as claimed in the '963 application. Also here, the presence of 1.5 wt. % NH<sub>4</sub>Cl dry on dry adhesive had limited influence on pH (see results in graph 1 and graph 2).
- Tinkelenberg et al does not explicitly or inherently disclose the compositions as claimed in the '963 application.

7. I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Respectfully Submitted,

May, 13, 2006

Date Signed

Karel F.H. BONEKAMP